Chemical Heterogeneity in Wildfire Plumes: Implications for Air Quality Models & Satellites

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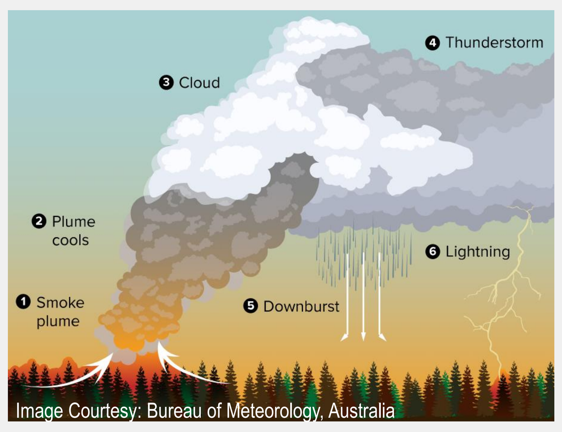
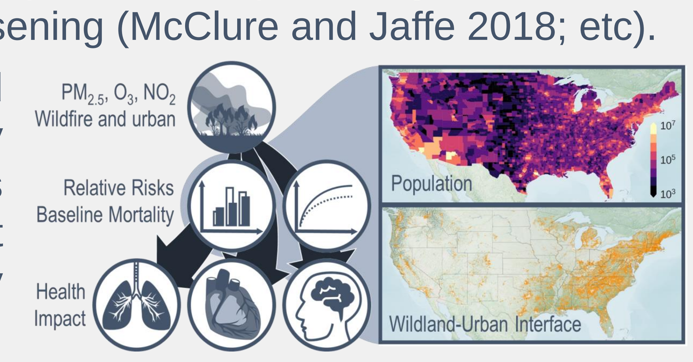
• Background •

- Wildland fires: a natural phenomenon, posing costly risks to human health and properties. With record high heats & severe drought, wildfires have becoming a growing concern in the U.S.

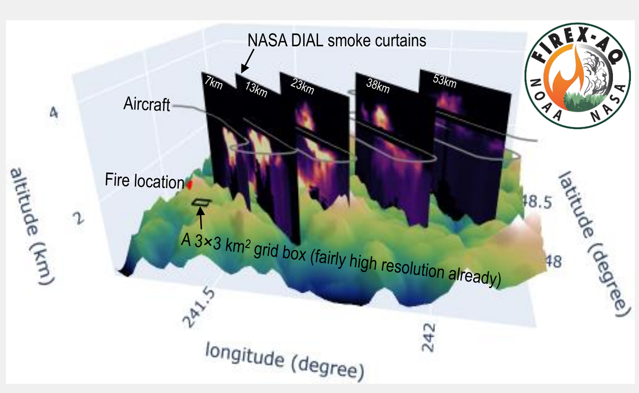


- Complicated entanglement between physics & chemistry: Fire-induced turbulence affects meteorology. Chemical processes produce many pollutants, e.g., O_3 and fine particulate matters ($PM_{2.5}$).
- Efforts to improve air quality over the past decades show promising trends, except in wildfire-prone regions where air quality has been worsening (McClure and Jaffe 2018; etc).
- Due to the broad impacts on air quality and climate, wildfires are a vital component in modern air quality & climate models.



• Challenges & Motivations •

- It remains challenging to represent wildfires in models. Main reason: model grids are often too coarse; subgrid parameterizations are often very problematic (e.g., plume rise).



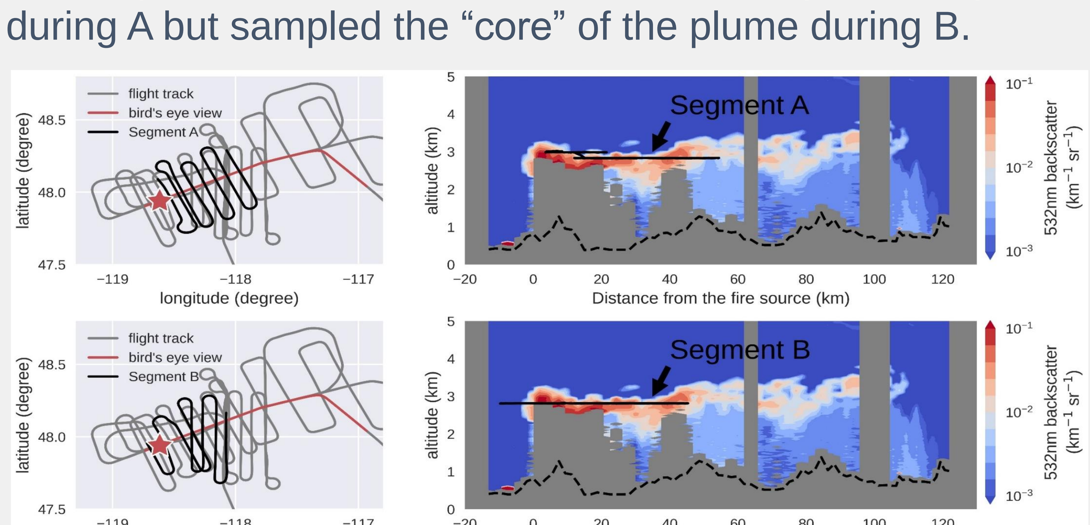
- As a result, physical and chemical processes in early stage of wildfire plumes (under-sampled too) cannot be explicitly resolved in current air quality / chemistry climate models.
- **Motivation:** Plume dynamics and the impacts on chemical evolution in the early stage of a large wildfire: Williams Flats fire (3 August 2019). This fire was extensively sampled during the NOAA/NASA FIREX-AQ field campaign.

• Large Eddy Simulation / Chemistry •

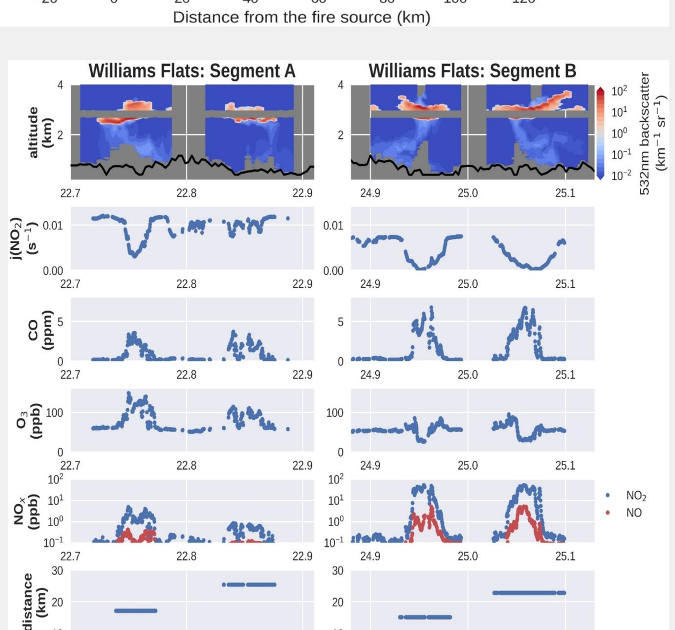
- A high resolution Large Eddy Simulation (LES) model (Moeng et al. 2017) coupled with simple yet representative chemistry is used in this work.
- Idealized LES in the WRF package. Driven by sounding profiles generated from a mesoscale (12 km) WRF-Chem simulation. LES domain size: 22 km × 22 km × 8 km. LES grid resolution: 100 m. Time step: 1 second.
- Chemical mechanism: O_3/CO/NO_x/VOCs + BC/OC (inert aerosols). Condensed largely based on MOZART T1.
- Photolysis and aerosol impacts on radiation: FTUV.
- Fire source characteristics: emissions of NO_x, CO, VOCs, BC, and OC taken from FINN2 then tuned until reasonable agreement is archived between airborne observations and model outputs. Sensible heat flux approximated from GOES-16 FRP products. Plume rise is explicitly resolved.

• Lidar Revealed Plume Structure •

- Airborne Lidar (NASA DIAL) revealed the plume vertical structure. The semi-Lagrangian sampling stage consisted of two segments: the aircraft skimmed the top of the plume during A but sampled the "core" of the plume during B.

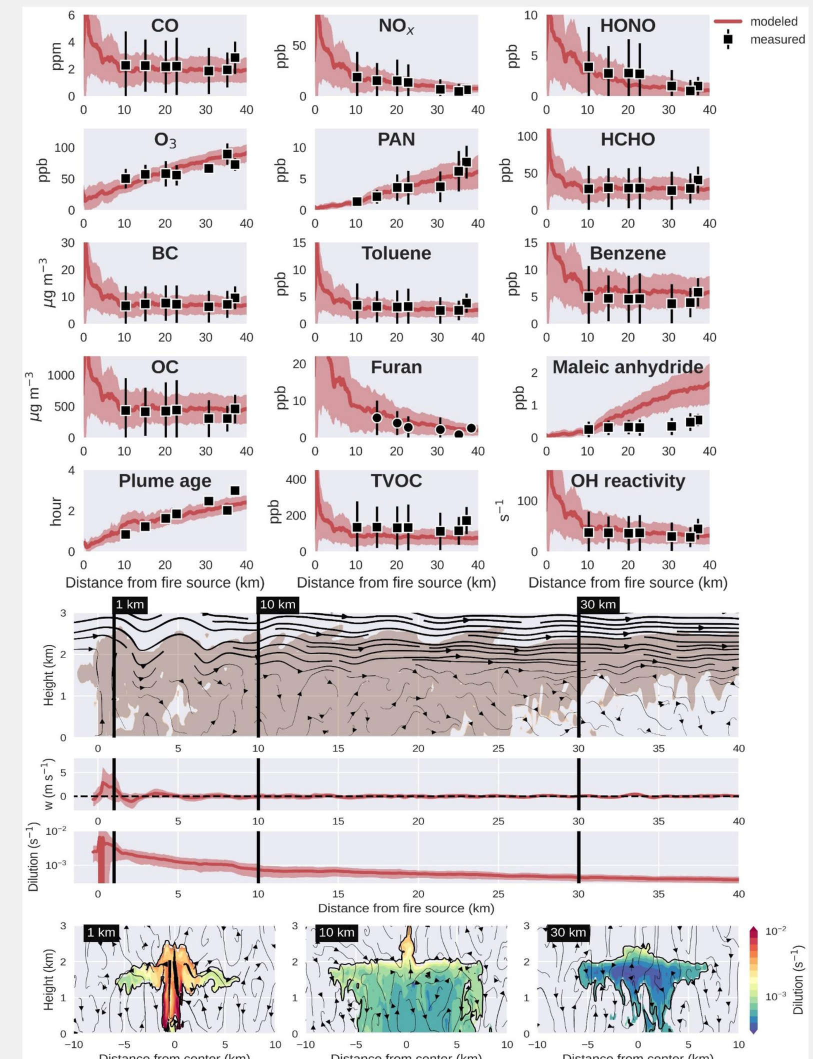


- In Segment B, j-values are greatly reduced at the center of the transect, where O_3 is severely titrated by NO. It can then be inferred the aircraft flew through the plume core during Segment B, which was therefore mainly used for model evaluation.



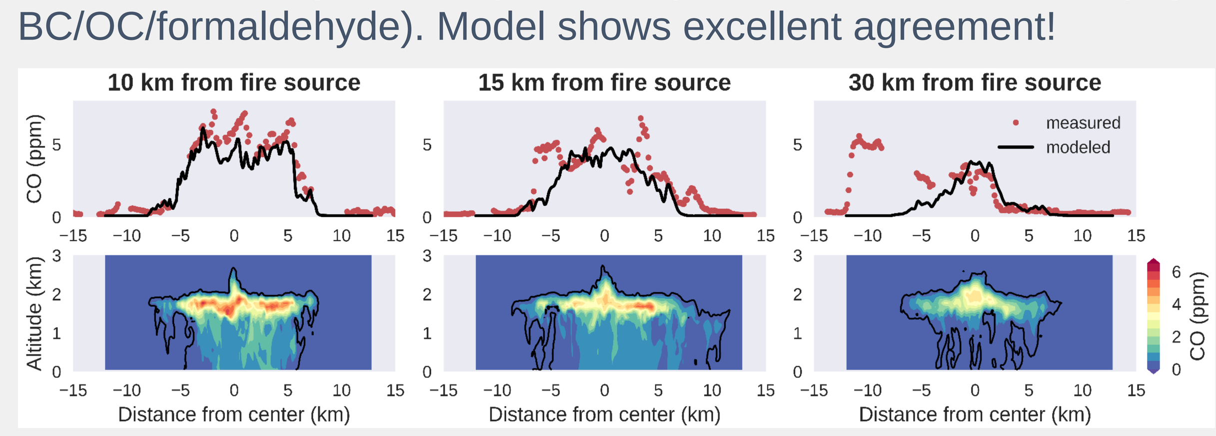
• Model Overview •

- 1-Hz data collected in each transect during was averaged and compared to the modeled at the same distance downwind. Model plume age and dilution tracked by two tracers (one inert, one with 1-hour lifetime). "Measured" plume age is based on wind data.
- Plume dynamics: strong heat release leads to rapid plume rise, producing downdrafts & small circulations near the plume, resulting in rapid dilution & entrainment.

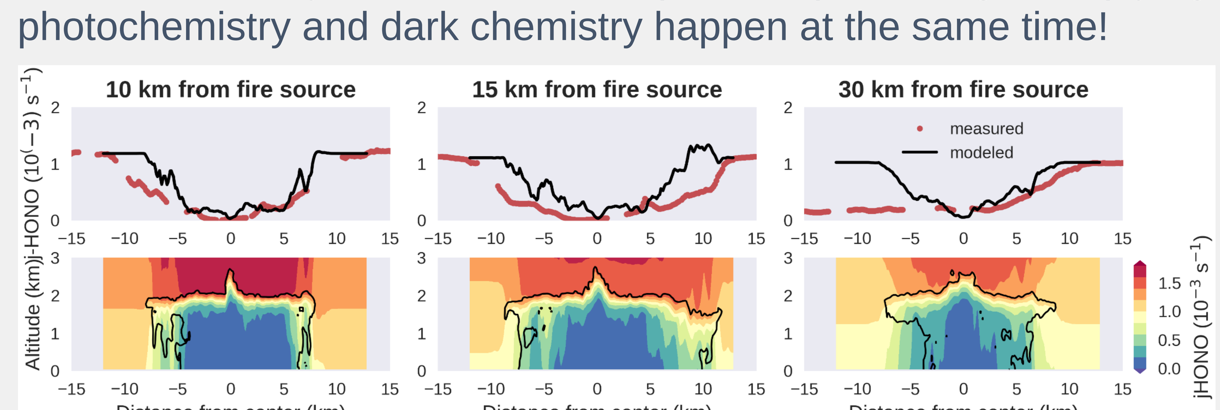


- Plume Chemical Heterogeneity -

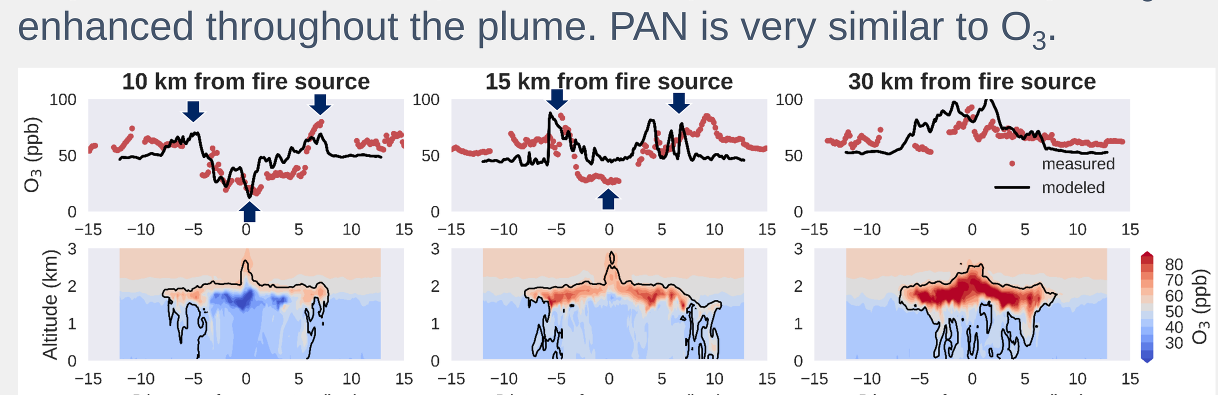
- Cross-transect variability of CO: peaked at plume center & reduced at edges. Many other pollutants are similar (e.g., BC/OC/formaldehyde). Model shows excellent agreement!



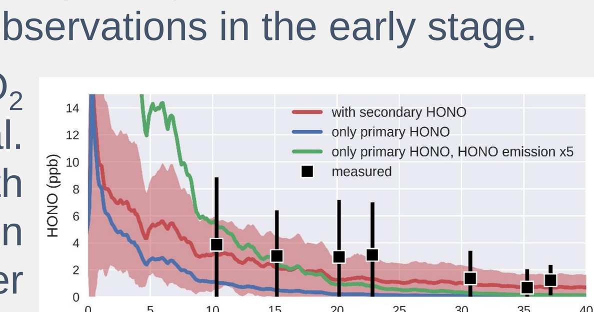
- J-values are suppressed at the center, due to the large amount of aerosols. Photolysis is faster at the plume top and edges, implying photochemistry and dark chemistry happen at the same time!

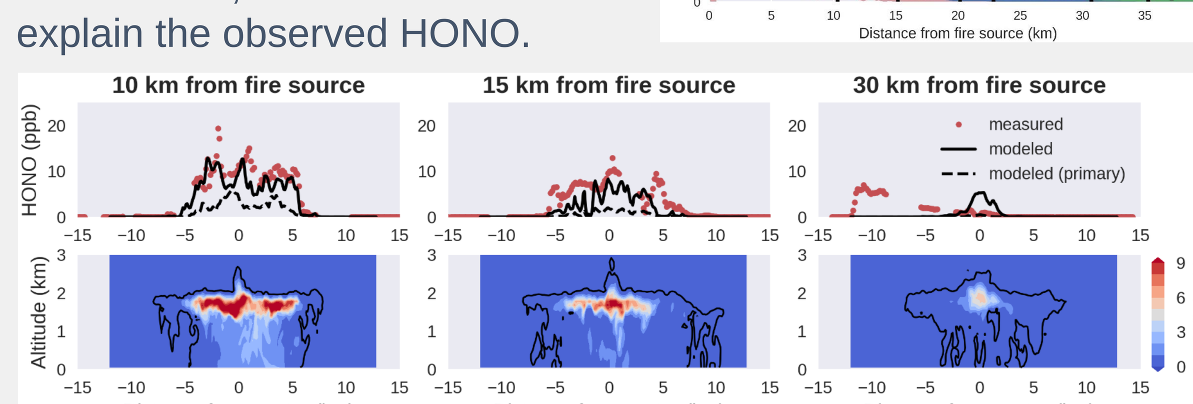


- O_3 is complicated! In the early stage, O_3 is severely suppressed at the dark center due to NO-titration, but may be enhanced at the edges due to active photochemistry. In the later stage O_3 is enhanced throughout the plume. PAN is very similar to O_3.

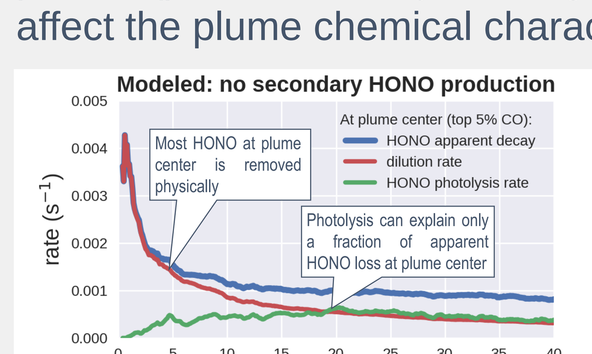


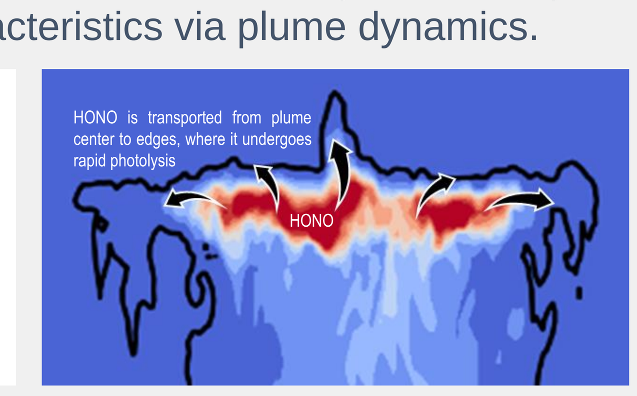
- HONO is a key oxidant in wildfire plumes, which can be directly emitted. HONO undergoes rapid photolysis. Modeled HONO shows excellent agreement with observations in the early stage.
- HONO may be produced from NO_2 update on aerosols (Ammann et al. 2013 and references therein). With a heterogeneous HONO formation on aerosols, the model can better explain the observed HONO.





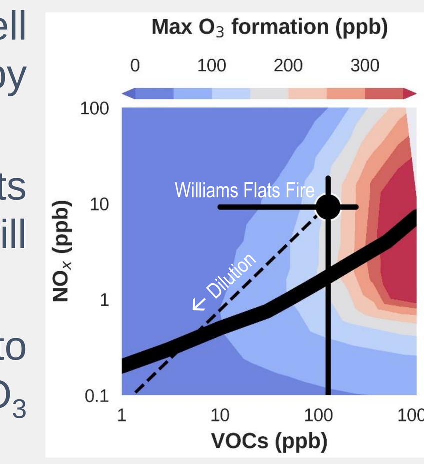
- HONO is rapidly killed at the edges but "protected" at the center! How fast HONO is killed is limited by how fast HONO can be transported from the center to the edges → bottleneck effect.
- Take-home: Different chemistry happening in different parts of the plume (photochemistry at edges, dark chemistry at center) all affect the plume chemical characteristics via plume dynamics.



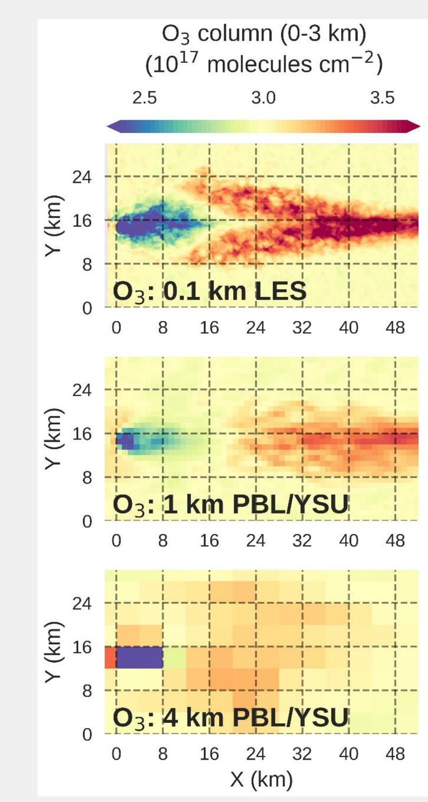


• Implications for Big Models •

- O_3 chemistry is highly nonlinear. It's well documented that O_3 formation is affected by VOCs and NO_x (duh).



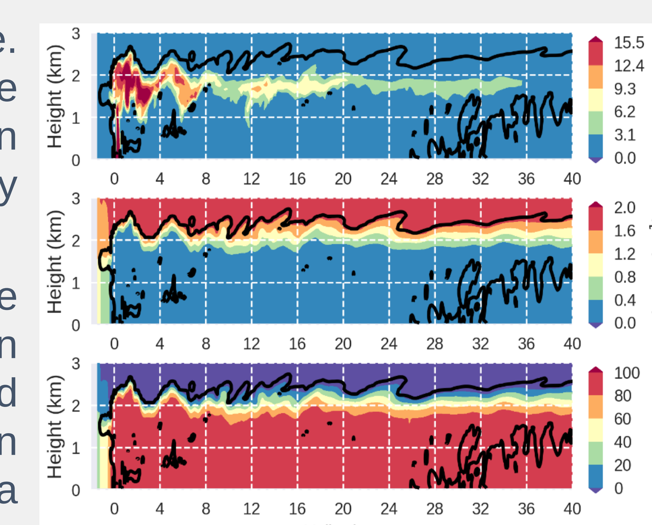
- In models with coarse resolutions, pollutants from point sources/small area sources will be immediately diluted: numerical dilution.
- Dilution, numerical or otherwise, will lead to a shift in chemical regime & bias in O_3 formation (e.g., EKMA plot).
- This effect is further demonstrated by showing the O_3 column (0-3km) in models with various resolutions. Chemistry is kept identical (emissions were regridded accordingly using conservative approach).



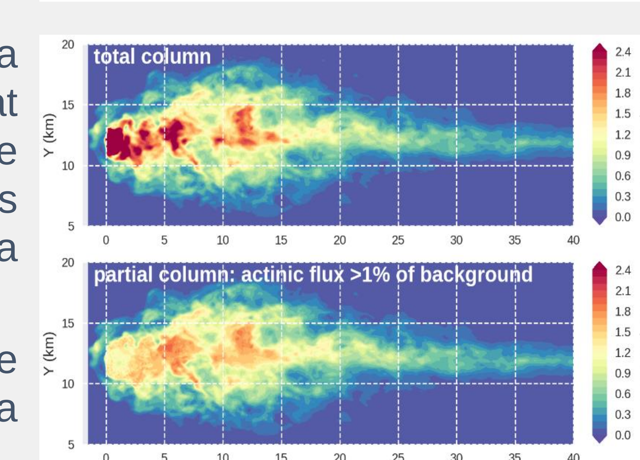
- As shown, if take the 0.1km LES as a benchmark, the 1km model (YSU) barely captured the O_3 characteristics, but the 4km model does not, due to bias introduced by numerical dilution.
- Even with "perfect" emissions and chemistry, the impacts of small wildfires (most of them really) on O_3 will be underestimated in models with coarse resolutions. → Need higher spatial resolution for air quality models!

• Implications for Satellite Retrievals •

- Take HONO for example. HONO is depleted at plume top, while satellite relying on UV/vis (Theys et al. 2020) only sees the top of thick plumes.



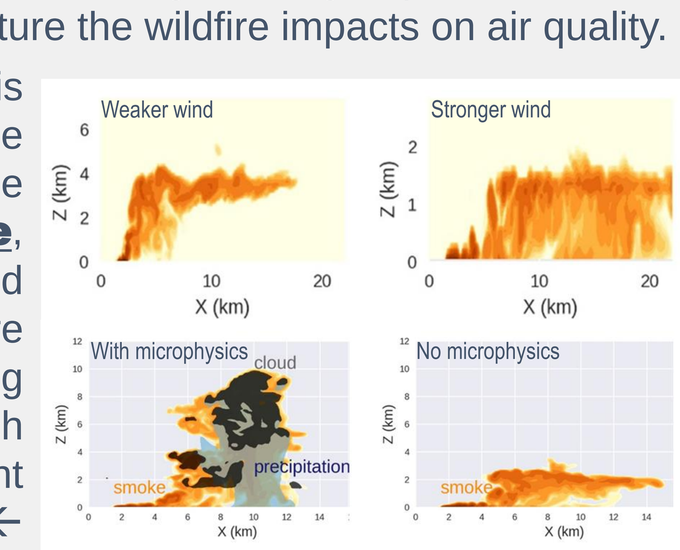
- Over the course of plume transport, light extinction inside the plume is reduced due to dilution, so satellite can see deeper → potentially a change in sensitivity?
- See demo on the right: With a less sensitive sensor that retrieves a partial column, the decay of HONO column is partially compensated by a change in sensor sensitivity.



- Satellite retrievals often use modeled plume profiles as a priori, which is problematic.

• Conclusions & Outlook •

- Chemical characteristics of wildfire plumes is highly complex, affected by both photochemistry (edges/top part of plume) and dark chemistry (interior/below) as well as plume dynamics.
- OH radicals formed from HONO drives the oxidation. HONO may be produced on from heterogeneous reactions on aerosols.
- Model resolution affects chemical regime! High spatial resolution (e.g., 1km) is needed to capture the wildfire impacts on air quality.
- Future outlook: we use this idealized LES to explore one of the weakest links in the chain, **plume rise**, targeting entrainment, wind shear, as well as moisture processes, utilizing advanced AI techniques such as random forest, gradient boost tree, neural network ← working in progress!



• References & Acknowledgements •

➢ Wang et al. (2021) *J. Geophys. Res. Atmos.* doi: 10.1029/2021JD035203
➢ McClure and Jaffe. (2018) *PNAS.* doi: 10.1073/pnas.1804353115
➢ Moeng et al. (2007) *Monthly Weather Review.* doi: 10.1175/MWR3406.1
➢ Ammann et al. (2013) *Atmos. Chem. Phys.* doi: 10.5194/acp-13-8045-2013
➢ Theys et al. (2020) *Nature Geosci.* doi: 10.1038/s41561-020-0637-7

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